THP5 Investigation of the Microscopic Parameters Governing the photorefractive effect in $KTa_{1-x}Nb_xO_3$: Cu,V

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We perform a microscopic analysis of the photorefractive effect in $KTa_{1-x}Nb_xO_3$:Cu,V. We have independently measured all relevant parameters which enter into the Kukhtarev model of the photorefractive effect. Cu is found to be the partially filled donor level responsible for the photorefractive effect. The total Cu concentration was determined from electron microprobe analysis to be $1.8(10^{19})cm^{-3}$. A series of oxidation and reduction treatments were used to alter the concentration of Cu^{1+} and Cu^{2+}. The concentration of Cu in each valence state after each heat treatment was determined using the relative magnitudes of the Cu^{1+} charge transfer band and the Cu^{2+} crystal field transition. The electron photoexcitation cross section at 514nm was determined to be $9.08(10^{-20})cm^{-2}$. The ratio of the mobility to electron recombination rate was determined from photoconductivity measurements to be $6.20(10^8)$ 1/(v-cm).

Oxidation and reduction treatments alter the photorefractive properties of metal oxide crystals by inducing changes in the valence state of dopant ions by altering the concentration of oxygen vacancies. The oxidation process is given by

$$Cu^{1+}+1/2V_O+1/4O_2 \rightarrow Cu^{2+} \quad (1)$$

where V_O represents an oxygen vacancy and O_2 an oxygen gas molecule. Charge neutrality is given by

$$4[Cu^{1+}]+3[Cu^{2+}]+n=2[V_o]+p \quad (2)$$

where n and p are the free electron and hole concentrations.
A mass action equation can be used to determine the concentration of the components of (1) in equilibrium

$$[Cu^{1+}][V_o]^{1/2}P_{O2}{}^{1/4}/[Cu^{2+}]=K\exp(dH/kT) \quad (3)$$

where P_{O2} is the partial pressure of oxygen, K is a constant, and dH is the change in enthalpy for the reaction. For each of the five heat treatments the temperature, P_{O2}, $[Cu^{1+}]$, and $[Cu^{2+}]$ are known. By neglecting the free carrier concentration relation (2) can be used to determine V_O. Using (3) we are able to determine dH=0.295eV and $K=2.83(10^{18})atm^{1/4}cm^{-3/2}$ (Fig. 1). dH is given by the band gap energy minus the energy below the conduction band of the Cu^{1+} level.

The above analysis allows us to determine the oxygen partial pressure and temperature necessary to control the Cu valence states (Fig. 2) and hence photorefractive properties of the KTN sample. We are thus able to vary and control the magnitude and response time for a given application. The theoretical and experimental dependence of these properties on heat treatments will be compared.

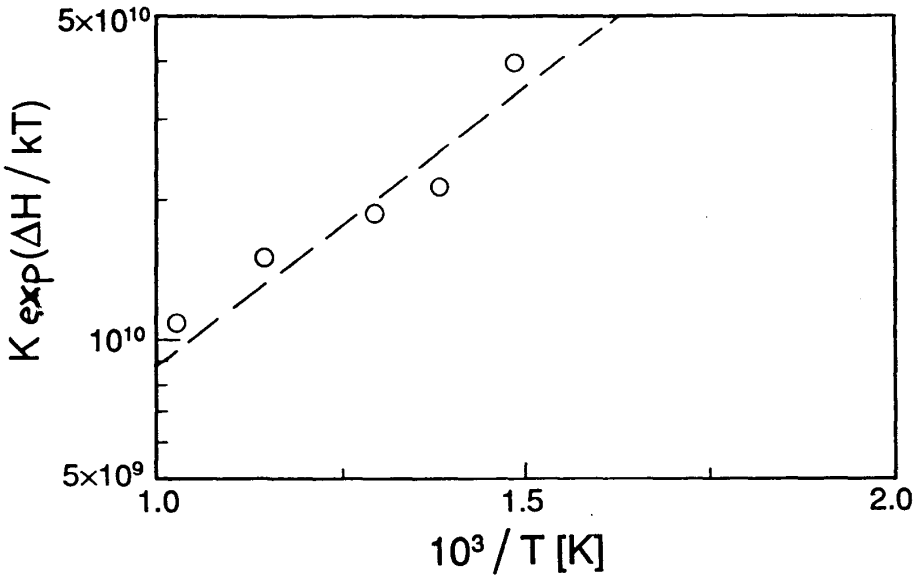

Fig. 1 Plot of the left hand side of the mass action equation (3) versus inverse processing temperature. Dashed line corresponds to dH=0.295eV and K=2.83EE18 $atm^{1/4}$ $cm^{-3/2}$.

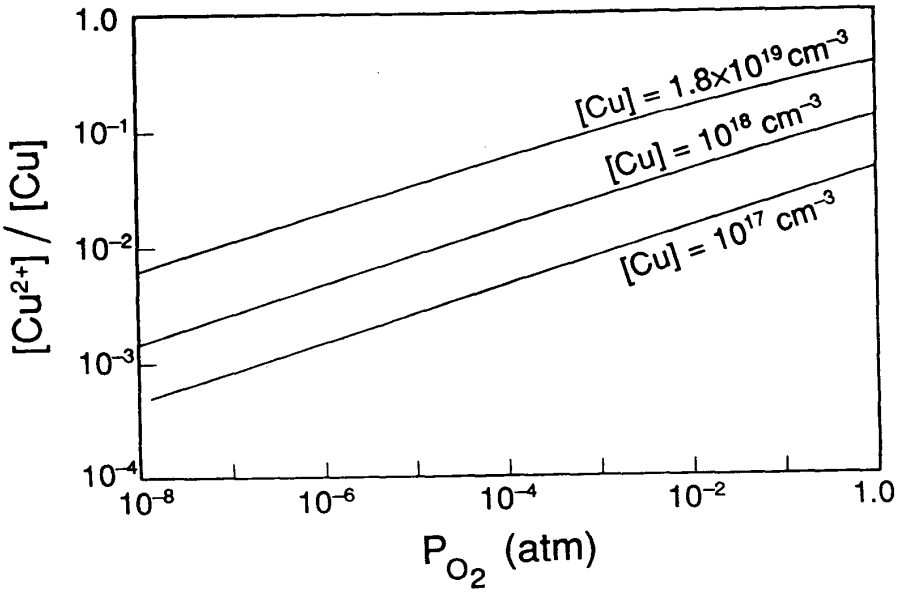

Fig. 2 Plots of the fraction of the Cu in the 2+ valence state versus partial pressure of oxygen for a 700C heat treatment. Plots for three doping levels are shown.